PATENT

#9/18 5/6/3

Practitioner's Docket No. 01-199

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

8 Mare application of:

Singleton, et al.

ation No.: 09/844,379

Group No.: 1754

Filed: 04/27/2001

Examiner: Jonas N. Strickland

For: ATTRITION RESISTANT FISCHER-TROPSCH CATALYST AND SUPPORT

Commissioner for Patents BOX AF Washington, D.C. 20231

TRANSMITTAL OF APPEAL BRIEF (PATENT APPLICATION--37 C.F.R. § 1.192)

- Transmitted herewith, in triplicate, is the APPEAL BRIEF in this application, with respect to the 1. Notice of Appeal filed on December 20, 2002.
- STATUS OF APPLICANT 2.

This application is on behalf of other than a small entity.

CERTIFICATION UNDER 37 C.F.R. §§ 1.8(a) and 1.10*

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^{*} Only the date of filing (§ 1.6) will be the date used in a patent term adjustment calculation, although the date on any certificate of mailing of transmission under § 1.8 continues to be taken into account in determining timeliness. See § 1.703(f). Consider "Express Mail Post Office to Addressee" (§ 1.10) or facsimile transmission (§ 1.6(d)) for the reply to be accorded the earliest possible filing date for patent term adjustment calculations.

3. FEE FOR FILING APPEAL BRIEF

Pursuant to 37 C.F.R. § 1.17(c), the fee for filing the Appeal Brief is:

Large entity

\$320.00

Appeal Brief fee due \$320.00

4. EXTENSION OF TERM

The proceedings herein are for a patent application and the provisions of 37 C.F.R. § 1.136 apply. Applicant petitions for an extension of time under 37 C.F.R. § 1.136 (fees: 37 C.F.R. § 1.17(a)(1)-(5)) for two months:

Fee: \$410.00

If an additional extension of time is required, please consider this a petition therefor.

Extension fee due with this request \$410.00

5. TOTAL FEE DUE

The total fee due is:

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\$410 00

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6. FEE PAYMENT

Charge the fee of \$730.00 to Deposit Account 06-0540.

A duplicate of this transmittal is attached for processing against the Deposit Account.

7. FEE DEFICIENCY

If any additional extension and/or fee is required, and if any additional fee for claims is required,

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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

APPLICANT:

SINGLETON, ET AL.

SERIAL NO.:

09/844,379

FILED:

04/27/2001

FOR:

ATTRITION RESISTANT FISCHER-TROPSCH CATALYST AND

SUPPORT

ART UNIT:

1754

EXAMINER:

JONAS N. STRICKLAND

CONFIRM. NO.: 1570

Commissioner for Patents Washington, D.C. 20231

APPELLANT'S BRIEF UNDER 37 C.F.R. § 1.192

Commissioner for Patents

BOX AF

Washington, D.C. 20231

> BOARD OF PATENT APPEALS AND INTERFERENCES ATTENTION:

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Please be advised that this brief is in furtherance of the Notice of Appeal filed in this case on December 20, 2002. The fees required under § 1.17 and any required petition for extension of time for filing this brief and fees therefor, are dealt with in the accompanying Transmittal of Brief.

This brief is submitted in triplicate.

This brief contains the items under the following headings required in accordance with 37 C.F.R. § 1.192(c) in the order set forth therein.

I. REAL PARTY IN INTEREST (37 C.F.R. § 1.192(c)(1))

The real party in interest in this application is:

Energy International Corporation 135 William Pitt Way Pittsburgh, PA 15238

The assignment of this application to Energy International Corporation was recorded in the United States Patent and Trademark Office at Reel 010000, Frame 0590 on May 21, 1999.

II. RELATED APPEAL OR INTERFERENCES (37 C.F.R. § 1.192(c)(2))

There are no other appeals or interferences known to Appellants, Appellants' legal representative, or the assignee which will directly affect or be directly affected by or have a bearing on the Board's decision in this appeal.

III. STATUS OF THE CLAIMS (37 C.F.R. § 1.192(c)(3))

This application is a divisional of U.S. Patent Application Serial No. 09/316,562, now U.S. Patent No. 6,262,132. Pursuant to a preliminary amendment filed with this divisional application on April 27, 2001, the claims originally contained in this application were claims 67-76 and 86-88. In an amendment filed July 10, 2002, claims 67-76 and 86-88 were canceled and replaced with claims 89-108. Claims 89-108 are presently pending in this application and are the subject of this appeal. Pursuant to 37 C.F.R. (37 C.F.R. § 1.192(c)(9), a copy of pending claims 89-108 is appended to this brief.

STATUS OF AMENDMENTS (37 C.F.R. § 1.192(c)(4))

On November 25, 2002, an amendment after final Office action was filed wherein claim 101 was amended to correct a typographical In an advisory action mailed December 10, 2002, the Examiner indicated that this amendment after final would be entered for purposes of appeal.

SUMMARY OF THE INVENTION (37 C.F.R. § 1.192(c)(5))

Applicants' claims 107 and 108 call for a y-alumina catalyst support material having improved attrition resistance. (9:16-19; 43:3-14; and 44, Table IV). Similarly. Applicants' claims 101-106

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call for a y-alumina catalyst support having improved attrition resistance for Fischer-Tropsch hydrocarbon synthesis. (9:16-19). Applicants' claims 89-100 call for a cobalt-on-y-alumina catalyst produced by depositing cobalt on the inventive, more attrition resistant y-alumina material. (7:11-17 and 21:9-10).

Applicants' new attrition resistant y-alumina support is unexpectedly produced by an inventive acid treatment process wherein the attrition resistance properties and characteristics of the y-alumina material itself are changed. In contrast to the prior art reference cited by the Examiner in this case, Applicants' invention therefore does involve or require the formation of any type of layered composite material wherein, rather than changing the properties and characteristics of the γ -alumina material itself, the y-alumina material must be encased within a relatively thick layer of a more attrition resistant, non-y-alumina material. (9:16-20; 43:3-14; and 44, Table IV). Further, the improved attrition resistance of the inventive y-alumina support material does not result from the addition of any type of catalytic material to the support. Rather, Applicants' claims clearly state that the inventive treatment of the y-alumina material with an acidic aqueous solution occurs prior to adding any catalytic material to the y-alumina. (9:10-20; 21:12-17; and 43:3-9).

The acidity level and the amount of the acidic aqueous solution employed to produce the inventive \u03c4-alumina support

material will preferably be effective for increasing the attrition resistance of the y-alumina support material for Fischer-Tropsch hydrocarbon synthesis in slurry bubble column reactors and other high agitation reaction systems. (7:11-17; 9:6-9; 22:12-18; 23:18-22; 43:3-14). The acidic aqueous solution preferably has a pH of not more than 5 and most preferably has a pH in the range of from about 3 to about 1. (43:12-14). The acidic aqueous solution is preferably formed using nitric acid and most preferably consists essentially of nitric acid and water. (43:4-5).

The γ -alúmina support material treated in accordance with the present invention is preferably a material which has been formed by calcining boehmite, preferably spray-dried synthetic boehmite, at a temperature and for a time effective to convert at least most of the boehmite material to a γ -alumina support material. (13:13-15; 13:17-19 and 43:3-4). The boehmite material is preferably calcined at a temperature in the range of from about 350°C to about 700°C (most preferably about 500°C) and will preferably have an average particle size of not more than 90 microns. (13:19-21 and 43:3-9).

Following the treatment of the γ -alumina support material with the acidic aqueous solution and prior to adding any catalytic material thereto, the treated γ -alumina material will preferably be re-calcined. This re-calcining step will preferably be conducted at a temperature such that at least most of the γ -alumina support material remains in γ -alumina form. The treated γ -alumina support

material will most preferably be re-calcined at a temperature of about 350°C. (43:6-8).

Applicants' inventive, more attrition resistant γ -alumina material called for in claims 107, 108, and 101-106 can generally be used for any purpose and is not limited to the manufacture of cobalt-on-alumina catalysts and/or to use in Fischer-Tropsch reaction systems. Claims 89-100, on the other hand, call for a cobalt-on-alumina catalyst having improved attrition resistance for Fischer-Tropsch hydrocarbon synthesis wherein, after being treated with an acidic aqueous solution and preferably also after recalcining, an effective amount of cobalt is deposited on the inventive γ -alumina support material. (18:5-17 and 19:1-9).

VI. Issues (37 C.F.R. § 1.192(c)(6))

Applicants' claims 89-108 stand rejected under 35 U.S.C. § 102(b) as anticipated by or, in the alternative, under 35 U.S.C. § 103(a) as obvious in view of U.S. Patent No. 5,037,791, issued to Comolli, et al. Pursuant to the Examiner's Advisory Action mailed December 10, 2002, it is Applicants' understanding that any and all other rejections of Applicants' claims have been overcome.

In his initial rejection of claims 89-108 in view of Comolli, et al., the Examiner stated that:

Comolli, et al. discloses having a cobalt metal deposited on a γ -alumina support, which exhibits reduced attrition losses (col. 1, lines 38-40; col. 2, lines 64-col. 3,

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line 4). The catalysts may be used in hydrocarbon production (col. 6, line 9).

The Examiner recognizes claims 89-108 as product-by-process claims. The Patent Office bears a lesser burden of proof in making out a case of prima facie obviousness for product-by-process claims. Once a rationale tending to show that the claimed product appears to be the same or similar to that of the prior art, although produced by a different process, the burden shifts to applicant to come forward with evidence establishing an unobvious difference between the claimed product and the prior art product. In re Marosi, 710 F.2d 798, 802, 218 USPQ 289, 292 (Fed. Cir. 1983).

Subsequently, in the advisory action mailed December 10, 2002, the Examiner stated that:

Comolli, et al. (USPN 5,037,791) discloses a cobalt metal deposited on a y-alumina support, which exhibits approved attrition loss wherein the process includes an acidic treatment. While Comolli, et al. teaches having a carbon layer, the instant claims are not limited to exclude a carbon layer. Furthermore, does not [sic] matter when the acidic treatment is performed within the process, as long as Comolli, et al. teaches wherein an acidic treatment is used in the process for producing a $\gamma\text{--}$ alumina support having improved attrition loss as well as improved catalytic performance. The applicant must come establishing an with evidence unobvious difference between the claimed product and the prior art product.

In view of the Examiner's remarks, Applicants believe the issues presented for this appeal are as follows:

- 1. Does the Comolli, et al. patent fail to disclose or suggest elements and limitations called for in Applicants' claims?
- 2. Has the Examiner failed to show that the Comolli, et al. product reasonably appears to be either identical to or

only slightly different from the product claimed by applicants?

3. Even assuming that the answer to the second issue is no, are there unobvious differences between the claimed product and the Comolli, et al. product.

VII. GROUPING OF THE CLAIMS (37 C.F.R. § 1.192(c)(7))

This application presently comprises the following three groups of claims:

Group I: Claims 89-100

Group II: Claims 101-106

Group III: Claims 107 and 108.

Pursuant to 37 C.F.R. § 1.192(c)(7), Applicants hereby state and affirm that all of the claims within each of Groups I, II, and III are separately patentable and that none of the claims within any of these groups stand or fall together.

VIII. ARGUMENT (37 C.F.R. 1.192(c)(8))

Applicants respectfully submit that Comolli, et al. neither disclose nor suggest several key features called for in Applicant's claims. Applicants further respectfully submit that, based upon a thorough reading of the Comolli, et al. patent, there is no reasonable basis for the Examiner's assertion that the Comolli, et al. product is either identical to or only slightly different from

Rather, it is

readily apparent that there are unobvious and unexpected

the product called for in Applicants' claims.

differences between the claimed product and the product disclosed

by Comolli, et al.

In complete contrast to the product and requirements of Applicants' claims, Comolli, et al. neither disclose nor suggest any procedure or process whereby the attrition resistance of any Y-alumina material itself is improved. Rather, Comolli, et al. teach only the production of a layered composite catalyst material formed by depositing a relatively thick carbon layer on a support material. The carbon layer must comprise from 5 to 40 weight percent of the composite.

Although Comolli, et al. teach that their resulting composite material has greater attrition resistance than the underlying support material alone, the purported improvement in attrition resistance does not result from any change in the properties or characteristics of the underlying support material itself. Rather, any difference between the attrition resistance of the Comolli, et al. composite and the underlying support material is due solely to the presence of the hard outer carbon coating.

In addition, in further contrast to the requirement in Applicants' claims that Applicants' acidic solution treatment be performed prior to adding any catalytic material to the support, the carbon layer of the Comolli, et al. catalyst apparently

constitutes a catalytic material which is activated by partial oxidation, pyrolysis, or reduction. (See Abstract).

Thus, contrary to the actual teachings of Comolli, et al., the attrition resistance improvement provided by Applicants' invention (a) involves a direct change in the characteristics and properties of a γ -alumina material itself, (b) does not involve or require any type of hard coating on the exterior of the γ -alumina material, (c) does not involved or result from the deposition of any type of catalytic metal or other catalytic material, (d) exists in the γ -alumina material prior to and without the deposition of any coating and/or catalytic material, and (e) is in no way limited, with respect to Applicants' claims 101-108, to use in cobalt catalysts.

In further contrast to the properties and characteristics of the layered composite taught by Comolli, et al., Applicants note that the improvement in attrition resistance provided by Applicants' invention is attained without any reduction in the surface area and pore volume of the γ -alumina material. Comolli, et al., on the other hand, admit that their carbon coating process significantly reduces both the surface area and pore volume. (See FIG. 2 and col. 8:14-30).

In addition to the above, Applicants are also at a loss as to the basis for the Examiner's new contention that Comolli, et al. somehow teach the use of an acid treatment in the process of producing a γ -alumina support having improved attrition loss. In

the Background of Invention section of the Comolli, et al. patent, at col. 1, lines 56-67, Comolli, et al. mention a spent catalyst regeneration procedure wherein carbon laydown and metal deposits can be removed, at least to some extent, by treating spent catalysts with dilute acid solutions. However, there is no indication or suggestion that such treatments can or should be used for regenerating spent catalysts having γ -alumina supports or that such treatment could conceivably reach or have any significant effect on the underlying support material of the caked spent catalyst. Further, the purported use of "dilute acid solutions" in such regeneration processes does not appear to indicate or suggest the use of aqueous acid solutions, particularly nitric acid solutions, having pH levels within the ranges called for in Applicants' claims.

To Applicants' knowledge, the only other instance in the Comolli, et al. patent where the use of an acid is mentioned is in Example 2 at col. 7, lines 13-20. In Example 2, Comolli, et al. merely indicate that, in one instance, an acid was apparently used for etching the outer carbon coating of a Comolli, et al. composite. However, there is no indication or suggestion that (a) such acid material could affect or even reach the underlying support material, (b) the acid material was applied in aqueous solution form or (c) the acid material was of a type and acidity level which, if applied in accordance with Applicants' claimed

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invention, would provide the unexpected attrition resistance improvement discovered by Applicants. In addition, it is also unclear as to whether the carbon coated, acid etched catalyst referred to in Example 2 of the Comolli, et al. patent included a gamma state alumina support material.

Finally, concerning Applicants' statement that all of Applicants' pending claims are separately patentable and that none of Applicants' claims within any of Groups I, II, or III stand or fall together, Applicants note that Comolli, et al. neither disclose nor suggest any of the various aqueous acidic solution compositions, acidic solution pH ranges or values, calcining steps and conditions, re-calcining steps and conditions, or preferred γ-alumina production techniques called for in Applicants' claims 89-108. Moreover, as indicated throughout the specification and examples of the application, the preferred conditions and requirements set forth in Applicants' various dependent claims provide further beneficial effects in the attrition resistance characteristics and properties of Applicants' inventive γ-alumina support material and catalysts.

Conclusion

In view of the above, Applicants respectfully submit that all of Applicants' pending claims 89-108 are in condition for allowance. Applicants therefore request that the Examiner's

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rejection of these claims be reversed and that all of claims 89-108 be allowed.

Respectfully submitted,

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APPENDIX OF CLAIMS IX.

Text of the claims involved in this appeal, including all entered amendments are as follows:

- A cobalt-on- γ -alumina catalyst having improved attrition resistance for Fischer-Tropsch hydrocarbon synthesis, said cobalton-y-alumina catalyst being produced by a method comprising the steps of:
 - calcining a boehmite material at a temperature and for a (a) time effective to convert at least most of the boehmite material to a y-alumina support material;
 - treating said y-alumina support material with an acidic (b) aqueous solution; and then
 - (c) forming said cobalt-on-y-alumina catalyst by depositing cobalt on said y-alumina support in an amount effective to catalyze said Fischer-Tropsch hydrocarbon synthesis,

wherein said y-alumina support material is treated in step (b), prior to depositing said cobalt in step (c), with said acidic aqueous solution at an acidity level effective for increasing the attrition resistance of said cobalt-on-y-alumina catalyst for said Fischer-Tropsch hydrocarbon synthesis.

- 90. The cobalt-on- γ -alumina catalyst of claim 89 wherein said γ -alumina support material is treated in step (b) with said acidic aqueous solution at an acidity level and in an amount effective for increasing the attrition resistance of said cobalt-on- γ -alumina catalyst for said Fischer-Tropsch hydrocarbon synthesis in a high agitation reaction system.
- 91. The cobalt-on- γ -alumina catalyst of claim 90 wherein said high agitation reaction system is a slurry bubble column reactor.
- 92. The cobalt-on γ -alumina catalyst of claim 90 wherein said acidic aqueous solution has a pH of not more than 5.
- 93. The cobalt-on- γ -alumina catalyst of claim 90 wherein said γ -alumina support material has an average particle size of not more than 90 microns.
- 94. The cobalt-on- γ -alumina catalyst of claim 90 wherein said acidic aqueous solution comprises water and nitric acid.
- 95. The cobalt-on- γ -alumina catalyst of claim 94 further comprising the step, after step (b) and prior to step (c), of recalcining said γ -alumina support material at a temperature such

that at least most of said γ -alumina support material remains in γ alumina form.

- The cobalt-on-y-alumina catalyst of claim 95 wherein said boehmite material is calcined in step (a) at a temperature in the range of from about 350°C to about 700°C.
- 97. The cobalt-on-y-alumina catalyst of claim 96 wherein said yalumina support material is recalcined in said step of recalcining at a temperature of about 350°C.
- The cobalt-on-y-alumina catalyst of claim 97 wherein said boehmite material is calcined in step (a) at a temperature of about 500°C.
- The cobalt-on-y-alumina catalyst of claim 94 wherein said acidic aqueous solution has a pH in the range of from about 3 to about 1.
- 100. The cobalt-on-y-alumina catalyst of claim 89 wherein said boehmite material is spray-dried synthetic boehmite.
- 101. A y-alumina catalyst support having improved attrition resistance for Fischer-Tropsch hydrocarbon synthesis, said y-

alumina catalyst support being produced by a method comprising the steps, prior to adding any catalytic material to said γ -alumina catalyst support, of:

- (a) calcining a boehmite material at a temperature and for a time effective to convert at least most of said boehmite material to a particulate γ -alumina support material having an average particle size of not more than 90 microns and then
- (b) treating said particulate γ -alumina support material with an acidic aqueous solution comprising water and nitric acid at a pH of not more than 5 and in an amount effective for increasing the attrition resistance of said particulate γ -alumina support material for said Fischer-Tropsch hydrocarbon synthesis and then
- (c) prior to adding any catalytic material to said particulate γ -alumina support material, recalcining said particulate γ -alumina support material at a temperature such that at least most of said particulate γ -alumina support material remains in γ -alumina form.
- 102. The γ -alumina catalyst support of claim 101 wherein said particulate γ -alumina support material is treated in step (b) with said acidic aqueous solution at an acidity level and in an amount effective for increasing the attrition resistance of said

particulate Y-alumina support material for said Fischer-Tropsch hydrocarbon synthesis in a high agitation reaction system.

- 103. The γ -alumina catalyst support of claim 102 wherein said high agitation reaction system is a slurry bubble column reactor.
- 104. The γ -alumina catalyst support of claim 101 wherein said acidic aqueous solution consists essentially of water and nitric acid and has a pH in the range of from about 3 to about 1.
- 105. The γ-alumina catalyst support of claim 101 wherein said boehmite material is a spray-dried synthetic boehmite material.
- 106. The γ -alumina catalyst support of claim 101 wherein said boehmite material is calcined in step (a) at a temperature in the range of from about 350° to about 700°C and said particulate γ -alumina support material is recalcined in step (c) at a temperature of about 350°C.
- 107. A γ-alumina catalyst support material having improved attrition resistance produced by a method comprising the steps of:
 - (a) treating a particulate γ -alumina material with an acidic aqueous solution comprising water and nitric acid and having a pH of not more than 5 and then

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(b) prior to adding any catalytic material to said particulate γ -alumina material, calcining said particulate γ -alumina material at about 350°C.

108. The γ -alumina catalyst support material of claim 107 wherein said acidic aqueous solution used in step (a) consists essentially of water and nitric acid and has a pH in the range of from about 3 to about 1.